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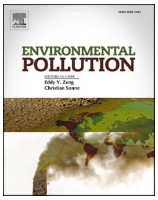

Effects of water chemistry and flow on lead release from plastic pipes versus copper pipes, implications for plumbing decontamination☆

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ABSTRACT

Despite being corrosion-resistant, plastic potable water pipes might accumulate heavy metals on their surface if they convey metal-contaminated tap water. This study examined the influence of water pH and flow conditions on lead (Pb) release from new and biofilm-laden potable water pipes to provide insights regarding decontamination. For this purpose, biofilms were grown onto new crosslinked polyethylene (PEX-A), high-density polyethylene (HDPE), and copper pipes for three months. Lead was then deposited onto the new and biofilm-laden pipes through 5 d exposure experiments under flow conditions. After that, lead release experiments were conducted by exposing the lead-accumulated pipes to lead-free synthetic tap water for 5 d, under both stagnant and water flow conditions. The metal accumulation study showed no significant difference in lead uptake by new pipes and their biofilm-laden counterparts under flow conditions. This could be attributed to the detachment of biofilms that have accumulated lead as water flows through the pipes. Water flow conditions significantly influenced the lead release from new and biofilm-laden water pipes. A lower water pH of 5.0 increased the release of lead from plastic pipes into the contact water, compared to pH 6.0 and 7.8. The greatest percentage of lead was released from biofilm-laden HDPE pipes (5.3%, 120 h) compared to biofilm-laden copper pipes (3.9%, 6 h) and PEX-A (3.7%, 120 h) and after exposure to lead-free synthetic tap water at pH 5.0, under stagnant conditions. On the other hand, under water flow conditions, the greatest lead release was found for new PEX-A pipes (4.4%, 120 h), new HDPE pipes (2.7%, 120 h), and biofilm-laden copper pipes (3.7%, 2 h).

1. Introduction

Despite the efforts of water distributors to deliver clean and safe drinking water to consumers, the chemical and microbiological quality of water might deteriorate as water is conveyed through the plumbing materials (Ghoochani et al., 2022; Hawes et al., 2017; Salehi, 2022). Potable water plumbing systems are associated with several public health concerns due to their potential to release heavy metals such as Pb, Cu, and Zn, and promote microbial growth in tap water (Ley et al., 2020; Proctor et al., 2020; Salehi et al., 2020). Over the last two decades, plastic potable water pipes such as high-density polyethylene (HDPE) and cross-linked polyethylene (PEX) pipes have been widely used to convey potable water within both water distribution systems and building plumbing to reduce cost and ameliorate drinking water quality concerns caused by corrosion of metallic pipes (Walsh, 2011). Although plastic potable water pipes have resolved historic corrosion concerns associated with metallic piping materials [e.g., galvanized iron, copper] (McFadden et al., 2011), they have also created additional concerns regarding their potential to leach organic compounds into tap water, which can sustain microbial regrowth (Salehi et al., 2018a; Wen et al., 2015). Migrating organic chemicals from plastic pipes can cause odor problems, promote microbial regrowth, and threaten the safety of tap water (Salehi et al., 2018a; Liu et al., 2017). Moreover, our prior research has shown that plastic materials could also serve as resting sites for the accumulation of heavy metals (Herath and Salehi, 2022; Salehi et al., 2018b; Hadiuzzaman et al., 2022). Previous research on lead fate in building plumbing systems has mainly focused on quantifying lead species in metallic pipes' surfaces, rather than polymers. Nevertheless, our field investigations have demonstrated that lead can also accumulate on the surface of PEX-A pipes (Salehi et al., 2017).

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Lead could be released into the tap water by corrosion of lead-containing plumbing materials such as lead service lines, solders, valves, galvanized iron pipes, brass fittings, and fixtures. The utilization of lead-bearing plumbing materials was prohibited in the United States since the implementation of Safe Drinking Water Act Amendment of 1986. However, its estimated by the U.S. Environmental Protection Agency (EPA) that there are still 6 to 10 million lead service lines in the U.S. The long-term lead release by lead service lines into the tap water could result in its accumulation onto the downstream potable water pipes in the buildings. The deposition of lead onto pipes can pose a public health threat when the deposited metals are released into the water under certain chemical, water flow, and/or microbiological conditions (DeSimone et al., 2020; EPA). Thus, decontamination of lead-accumulated pipes could be considered as an effective action to prevent the potential future chronic and acute exposure incidents. The study conducted by Huang et al. (2017) reported utilizing a biomass-derived ligand for cleaning the metal accumulated plastic water pipes that were removed from a residential building after one year of operation (Huang et al., 2017). However, to our knowledge, no study was found reporting the metal release from metal-accumulated plastic water pipes considering the interrelated roles of biofilm presence and water flow conditions.

Water pH and disinfectant residuals are known to be the major factors that influence the metals dissolution and release. Water chemistry fluctuations, especially reducing pH could result in dissolution of precipitated metal species that have been accumulated on the plastic surface (Lasheen et al., 2008). Moreover, shear forces caused by water flow can dislodge the metal species that deposited on pipe surface (Ahamed et al., 2020). Metal release can occur through ion exchange processes where metal ions are exchanged with the ions of similar charge from the aqueous solution. This mechanism is driven by the differences in ion concentrations and can be influenced by water pH and ionic strength (Huang et al., 2017). Wang et al. (2014) reported that heavy metal ions can associate with functional groups of hydroxyl (-OH) and carboxyl (-COOH) present on plastic surfaces through ion exchange process and then be released into water due to the disruption of equilibrium. The extent of ion exchange processes depends on a polymer's surface charge, functional groups, the ionic strength of water, and ionic concentration. Investigations on the influence of biofilms on metal release from pipes into the water are limited to metallic pipes (Lead; Pehlivan and Altun, 2007; Wang et al., 2014). Despite the critical role biofilm play in sequestering the heavy metals present in tap water (Wang et al., 2012), no systematic investigation has been conducted to describe how biofilm presence on plastic water pipes might impact metal release processes. One recent study showed that biofilm formation on the surface of polyethylene pellets significantly increases the amount of lead deposited onto them, but biofilm impact on lead release was not invesitigated (Ahamed et al., 2020).

Metal release from plastic potable water pipes has received less attention from researchers, despite the significant use of plastic plumbing materials for new constructions and its potential impact on tap water safety. Therefore, this study is conducted to examine the role of water chemistry and water flow conditions on lead release from new and biofilm-laden water pipes. The specific objectives are to (1) examine the lead release from new PEX-A and HDPE pipes under stagnant and flow conditions over time, (2) investigate the impact of biofilms on lead release from HDPE, PEX-A, and copper pipes, and (3) elucidate the effect of water flow and chemistry conditions on lead release from potable water pipes. We hypothesized that biofilm detachment under flow conditions and lower water pH promote the lead release from metal-accumulated pipes into the contact water.

2. Experimental

2.1. Materials

Crosslinked polyethylene type A (PEX-A) and high-density polyethylene (HDPE) pipes, with average diameters of 17 mm and 21 mm, respectively, and copper pipes with an average diameter of 19 mm, were purchased from a local hardware store (Memphis, TN, USA). Sodium hypochlorite (NaOCl) was purchased from RICCA (Arlington, TX, USA), and Pb ICP-MS standard solution (1000 ppm) was purchased from Thermo Fisher Scientific Inc (Waltham, MA, USA). Ultrapure Milli-Q™ (18MΩ*cm) treated water was used for all the experiments in this study. The composition of synthetic tap water used for all metal exposure and release experiments is shown in Table SI-1.

2.2. Biofilm growth

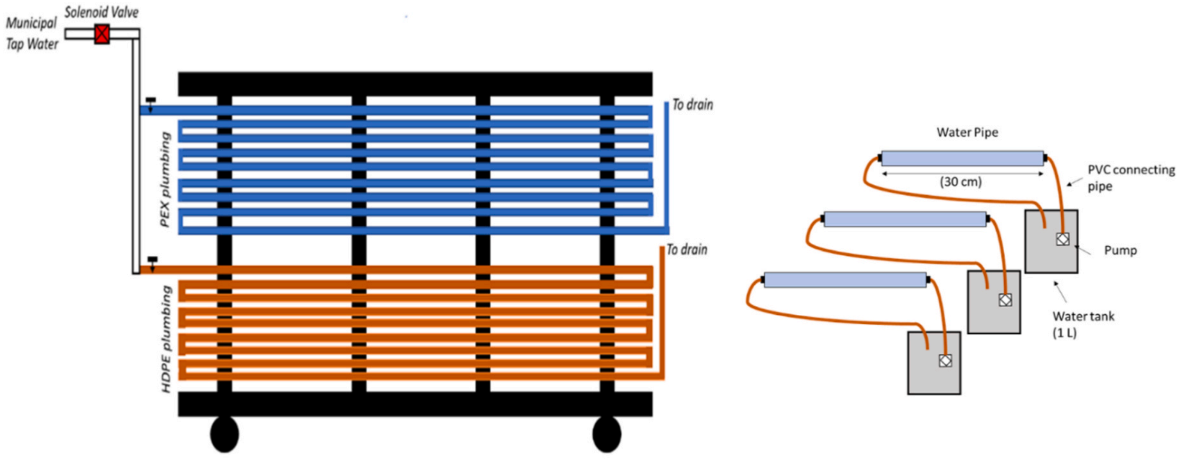
New PEX-A, HDPE, and copper pipes were first rinsed with tap water and then disinfected using 20 mg/L of NaOCl for 40 min to eliminate any previous microbial colonization. Biofilms were established on the pipes by running municipal tap water through a pipe loop at a flow rate of 2 ± 0.2 L/min for 90 d at 25 ± 1 °C, as shown in Fig. 1a. The setup comprised three independent closed pipe loops, each with a length of 305 cm. The biofilm growth process for the three types of pipes was not conducted simultaneously. Water flow was controlled automatically using a solenoid valve to achieve 16 h of flow and 8 h of stagnation daily. Tap water quality parameters, including temperature (PEX-A: 16.85 ± 0.8 $^{\circ}C$, HDPE: 17 ± 0.8 $^{\circ}C$, and copper: 17.5 ± 0.8 $^{\circ}C$), pH (PEX-A: 6.4 ± 0.4, HDPE: 6.1 ± 0.6, and copper: 6.1 ± 0.5), and total chlorine (PEX-A:1.2 ± 0.1 mg/L, HDPE: 1.2 ± 0.1 mg/L, and copper: 1.2 ± 0.1 mg/L), were measured, weekly. After 90 d of biofilm growth, the biofilm-laden pipes were cut into 30 cm segments to undergo subsequent metal deposition experiments under flow conditions. The chemical quality of tap water supplied by the local water utility is shown in (Salehi et al., 2021).

2.3. Biomass quantification

Representative 9 cm sections of pipes were placed into a sterile 50 mL centrifuge tube with 40 mL sterile 1x phosphate-buffered saline (PBS) solution. Total genomic DNA (gDNA) was extracted from these pipes using PureLink® Genomic DNA Mini Kit (Invitrogen, Carlsbad CA) following standard procedures. Adhered biofilms were liberated by physically scraping with sterile single-use stiff bristle plastic brushed for 5 min based on Chatterjee et al. (2012), with modifications. The liberated biofilms underwent centrifugation at 10,000×*g* for 10 min at 4 °C to pellet biofilms and supernatant was removed. Pellets were resuspended in 500 μL of molecular grade (DNase, RNase, and Proteinase free) water and transferred to extraction tubes. The extracted DNA was quantified using a Nanophotometer N60 (IMPLEN, Munich, Germany). The internal surface area of each pipe was determined using digital calipers with a resolution of one-tenth of a millimeter (Fisherbrand Traceable, Waltham, MA). Biofilm biomass (proxied by absolute quantification of copy numbers of the ribosomal DNA (16s) was evaluated by using droplet digital PCR (ddPCR). We quantitated number of copies of the V4 region of the 16S ribosomal DNA, which was then normalized to a per unit of surface area (cm^2) value. More information is provided in **SI-1**.

2.4. Lead deposition experiments

To study lead release from new and biofilm-laden water pipes, lead deposition experiments were first conducted under flow conditions. The new pipes were disinfected with 20 mg/L of NaOCl for 40 min prior to the lead deposition experiments, but the biofilm-laden pipes were subjected to the lead deposition, directly. The 30-cm sections of new and biofilm-laden water pipes were used in a small pipe loop for lead deposition experiments as shown in Fig. 1b. The ratio of aqueous lead



(a) (b)

Fig. 1. Schematics showing (a) pipe loops used for biofilm growth and (b) small pipe loop used for metal deposition and release experiments.

solution volume to the pipe length was selected as 1 L/ft. A small submersible water pump with a constant flow rate of 2.5 L/min (cross flow velocity of 11.0 m/min for PEX-A pipe, 7.2 m/min for HDPE pipe, and 8.8 m/min for copper pipe) was used to circulate the metal aqueous solution through the pipe loop. More information is provided in **SI-2**. The total mass of lead that accumulated on the pipes' surface after 5 d of lead deposition experiments was calculated using equation (1). In which $[Pb]_{in}$ is the initial lead concentration (μg/L), $[Pb]_{aq}$ is the concentration (μg/L) of the residuals lead left in the aqueous solution after 5 d, and $[Pb]_{rinsate}$ is the lead concentration (μg/L) in the rinsate of the pipe loop components, V_{aq} (L) is the volume of aqueous solution, and V_R (L) is the volume of rinsate. To determine the lead surface loading on pipes, the total mass of lead accumulated on pipe surface was divided by the surface area of the pipes, which were 0.016 m^2, 0.020 m^2, and 0.019 m^2 for 30 cm of PEX-A, HDPE, and copper pipe segments, respectively.

$$M(Pb)_{pipe} = ([Pb]_{in} \times V_{aq}) - \left([Pb]_{aq} \times V_{aq}\right) - ([Pb]_{rinsate} \times V_{aq}) \tag{1}$$

2.5. Lead release experiments

Lead release experiments were conducted under both water flow and stagnant conditions to gain a better understanding of how different water usage patterns can affect the lead release behavior from new and biofilm-laden water pipes. The experiments were conducted in triplicates. To investigate the lead release under flow conditions, the pipe segments were placed in a small pipe loop. Each end of the pipe segment was connected to two separate 61 cm long clear vinyl tubes, which were connected to the water tank. A small submersible pump was used to pump the water through each pipe segment with a constant flow rate of 2.0 L/min. In the lead release experiments, the assigned pipe segments with accumulated lead were exposed to lead-free synthetic tap water at a designated pH to investigate the impact of water chemistry on lead release behavior. For lead release experiments conducted under flow conditions, 5 mL water samples were collected after 5 min, 2 h, 6 h, 12 h, 24 h, 48 h, and 120 h. However, for the lead release experiments conducted under stagnant conditions, all water content of each pipe segment was acidified and then analyzed for lead quantification. The percentage of lead release was determined by dividing the mass of lead released from each pipe section by the total lead mass accumulated onto the same pipe section after each time interval. More information about statistical analysis is provided in **SI-3**.

2.6. Water quality measurements

The total Pb concentration in water samples was measured using a PerkinElmer Atomic Absorption spectrometer (AA400, HGA 900 Graphite Furnace). The instrument was calibrated with eight standards ranging from 0 μg/L to 100 μg/L. The limit of detection (LOD) was found to be 1.7 ± 0.4 μg/L. All standard curves had a coefficient of determination in the range of 0.98–0.99. Water pH was measured using a Fisherbrand™ accumet™ XL600 pH Meter. Hach Pocket Colorimeter™ II was used to measure the total chlorine residual (detection limit 0.1 mg/L).

3. Results and discussion

3.1. Lead (Pb) speciation in synthetic tap water

The lead speciation in synthetic tap water was determined using Visual MINTEQ 3.1 software. Fig. 2 shows the distribution of lead species in synthetic tap water with an initial lead concentration of 600 μg/L as a function of pH. The major ligands present in this aqueous system are OH^-, CO_3^{2-}, NO_3^-, SO_4^{2-} PO_4^{3-} and Cl^- which form various complexes such as $Pb(OH)_2$ (aq), $Pb(OH)_3^-$, $Pb(OH)^+$, $Pb(CO_3)_2^{2-}$, $Pb(CO_3)$ (aq), $Pb(NO_3)$ (aq), $PbSO_4$ (aq), and $Pb(SO_4)_2^{2-}$. At pH 8.2, only 3.2% of $[Pb]_t$ was dissolved, and 96.8% of $[Pb]_t$ was precipitated as $Pb(OH)_2$(s). Reducing the pH to 5.9 resulted in favorable conditions for the precipitation of $Pb_3(PO_4)_2$ (s) without the formation of $Pb(OH)_2$(s). As the pH decreases, the concentration of OH^- decreases, making $Pb(OH)_2$ less stable in the aqueous solution. At the same time, the presence of PO_4^{3-} enhances the formation of $Pb_3(PO_4)_2$ (s), which has a lower solubility compared to $Pb(OH)_2$ (s). In this study, lead accumulation experiments were conducted at pH = 7.8. At this pH, the dissolved lead species including Pb^{2+}, $PbCO_3$(aq), and $Pb(OH)^+$, accounted for only 10% of $[Pb]_t$, while 89.60% of $[Pb]_t$ was precipitated as $Pb_3(PO_4)_2$(s). However, by reducing the pH from 7.8 to 6.0, the percentage of dissolved lead species increased from 10% to 83.1% of $[Pb]_t$. Further decreasing the pH to 5.0 resulted in an increase in the percentage of dissolved lead species to 99.7% of $[Pb]_t$. Although the systems for lead accumulation and release experiments were closed, the pH of the aqueous solution may have changed. Thus, the speciation results found from the modeling may not be exactly representative of the experimental systems. However, it suggests that the reduction of water pH during the release experiments causes the dissolution of lead species that may have precipitated onto the pipe surface, subsequently leading to their release into the tap water.

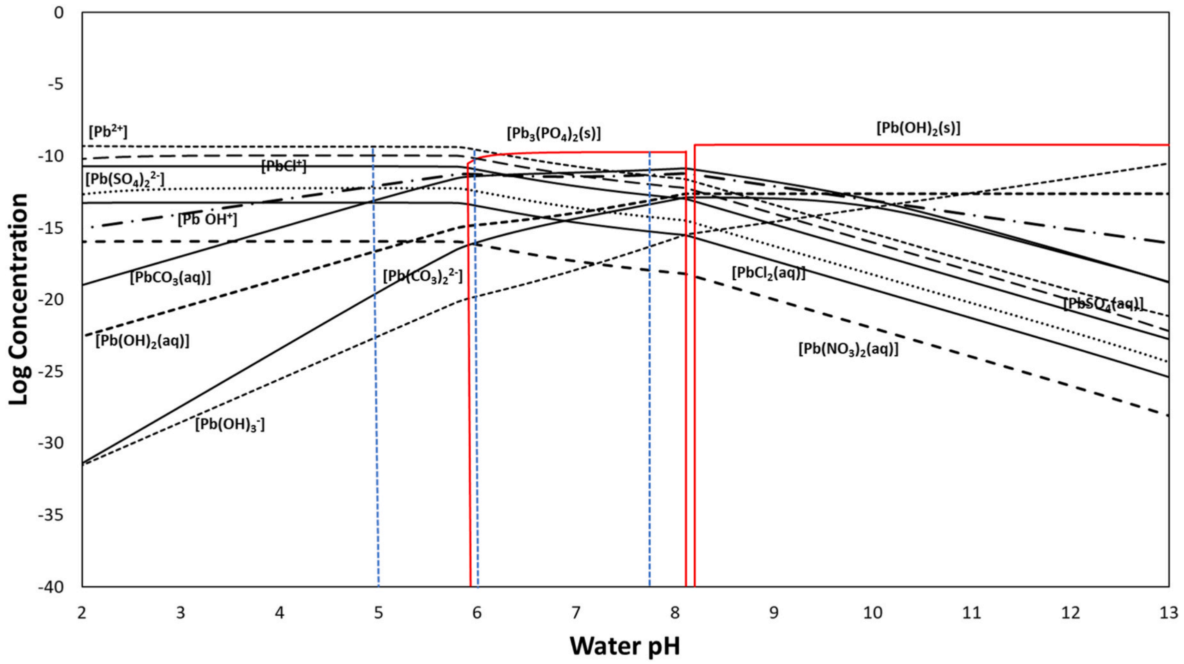


Fig. 2. Variations of Pb^{2+} species concentrations versus pH at $[Pb]_t = 600$ μg/L.

3.2. *Biofilm biomass quantification*

Biofilm biomass on the inner surface of the pipes was quantified for the representative pipe samples after three different stages of (i) biofilm growth, (ii) lead accumulation, and (iii) lead release. The results obtained for the representative samples showed that after 90 d of biofilm growth, there was higher biomass on PEX-A pipes than on HDPE and copper pipes. In particular, the surface loading of biomass accumulated on PEX-A pipes was 27,852,331 copies/cm^2, which was 8 and 17 times greater than that on HDPE (3,538,218 copies/cm^2) and copper (1,610,041 copies/cm^2) pipes' representative samples, respectively. This finding is consistent with literature that reported a greater biofilm biomass accumulation on plastic pipes compared to copper pipe. Specifically, it suggests that PEX-A pipes favor more biofilm growth than HDPE and copper pipes, likely due to surface roughness characteristics (Figure SI-1). Moreover, it has been reported that greater organic leaching from PEX-A pipes compared to HDPE pipes may have contributed to greater biofilm biomass (Salehi et al., 2021; Picone et al., 2021). On the other hand, copper pipes are known for their antibacterial characteristics, which could have contributed to their lower biofilm biomass accumulation (Connell et al., 2016; Gomes et al., 2020). Plastic materials are reported to rapidly colonize microbial contaminants in significantly higher densities than those observed for copper (Vargas et al., 2014). Our finding also agrees with Lehtola et al. (2005), that showed biofilm grows faster on polyethylene pipes than on copper pipes (Lehtola et al., 2005). Copper pipes' slower biofilm formation is attributed to the presence of copper ions, which exhibit antimicrobial properties and inhibit bacterial growth (Rogers et al., 1994; Straub et al., 1995; Learbuch et al., 2021). The literature suggested that the formation of biofilm on copper pipes requires more time than on plastic pipes, as copper can inactivate bacteria by attacking their respiratory enzymes or nucleic acids. On the other hand, the release of phosphorus from polyethylene plumbing materials has been suggested as the reason for their greater biofilm biomass compared to copper pipes (Lehtola et al., 2004).

Lead accumulation experiments were conducted for 5 d using a small pipe loop at an approximately similar flow rate to the one used for the biofilm growth process. The representative samples suggested that this process reduced the extent of biofilm biomass surface loading to less than 10% of its initial value for both PEX-A (748,751 copies/cm^2) and copper pipes (15,168 copies/cm^2), while more than 50% of the initial biofilm biomass remained on the HDPE pipe surface (1,893,343 copies/cm^2). Biomass release from the pipe surface during the lead accumulation experiments can occur through several processes. The physical disturbance of biofilm during the lead accumulation experiments can lead to biomass release. The shear stress created by water flow can cause the detachment of biofilm and its subsequent release into the water (Schwartz et al., 1998). Additionally, the toxicity of the lead accumulated onto the biofilm surface could have contributed to its detachment (Shen et al., 2015).

Biofilm biomass was also quantified in pipe samples after undergoing 5 d release experiments at pH 5.0 under stagnant and flow conditions. The results showed that under both conditions, biofilm regrowth likely occurred in PEX-A pipes, while the extent of biofilm biomass was reduced for HDPE pipes. The results obtained for the representative samples suggest that PEX-A pipes are more susceptible to biofilm regrowth as they are exposed to clean synthetic tap water compared to HDPE and copper pipes. It could be due to its surface morphology characteristics that promote the surface attachment of biomass, as shown in Figure SI-1. Studies have shown that the tendency of PEX pipes to leach microbial nutrients such as phosphates into the water leads to the promotion of biofilm formation, particularly in the weeks immediately following installation (Qi et al., 2021). Moreover, water pH may have impacted microbial regrowth. The release experiments were conducted at pH 5.0, which was lower than the pH of 7.8 that was used for the accumulation experiment. The effect of pH on bacterial attachment to the surface and biofilm formation can be organism-dependent (Lehtola et al., 2004).

The results suggest that stagnant conditions may increase the accumulation of biomass on pipe surfaces, particularly for PEX-A pipes. The biomass accumulations on PEX-A (2,323,068 Copies/cm^2) and HDPE (14,846 Copies/cm^2) were greater under stagnant conditions compared to the flow conditions, where the biofilm biomass on PEX-A and HDPE were 16,827,016 Copies/cm^2 and 274,304 Copies/cm^2, respectively. Under flow conditions, water flow might create shear forces on the pipe surface, which can physically detach the biomass from the pipe surface by overcoming the adhesive forces between the biofilm matrix and pipe surface, but additional investigations are needed to confirm. Additionally, the lack of water flow is likely to provide a more stable environment for microorganisms to grow and attach to the pipe surface. On the other hand, under water flow conditions, the movement of water can physically remove the biomass or available nutrients, thereby reducing the accumulation of biomass.

3.3. Lead (Pb) accumulation onto new and biofilm-laden water pipes

New and biofilm-laden water pipes were exposed to lead aqueous solution ($[Pb]_t = 600$ μg/L, pH = 7.8) for 120 h under flow conditions to accumulate lead onto pipes. The mass of lead accumulated on 1 ft of new pipes and biofilm-laden water pipes after 5 d exposure is shown in Figure SI-1. The mass of lead accumulated on 30 cm of new PEX-A, HDPE, and copper pipes was found as 596 μg, 594 μg, and 571 μg, and the mass of lead accumulated onto 30 cm of biofilm-laden PEX-A, HDPE, and copper pipes were found as 580 μg, 589 μg, and 568 μg, respectively. No significant difference (p-value >0.05) was found in lead accumulation onto the new pipes and their corresponding biofilm-laden pipes. This could be associated with the detachment of biofilm from the pipe surface that resulted in releasing previously sequestered lead into the contact water during the 5 d exposure experiments. The biofilm biomass quantifications also demonstrated that after lead accumulation experiments, there were 47% and 97% biomass reduction in the HDPE and PEX-A pipe surfaces, respectively. It should be noted that the biofilm growth process was conducted only for three months; this may have been attributed to its weak attachment to the pipe surface and subsequent detachment during the lead accumulation experiments. Furthermore, the lead accumulation experiments involved circulating a lead aqueous solution in the pipe loop for 5 d. So, after the initial saturation of surface sites and subsequent precipitation of lead species on available nucleation sites, this may have led to a distinction in lead surface loadings between new and biofilm-laden pipes.

The lead speciation results indicated that at pH 7.8, 89.60% of $[Pb]_t$ was precipitated as $Pb_3(PO_4)_2(s)$. This suggests that biofilm could have adsorbed lead species or have provided nucleation sites for the lead crystals to grow. However, as experiments were conducted on the flow conditions, shear stresses may have resulted in detachment of biofilms. Consequently, some of the surface-deposited lead in the biofilm may have been released into the contact water. Despite the greater percentage of biofilm detachment from the HDPE pipes compared to the PEX-A pipes, the reduction in lead uptake was about equivalent for both materials. This finding is similar to our recent study, which showed water flow conditions resulted in a lower lead accumulation onto biofilm-laden water pipes compared to new pipes. However, the extracellular polymeric substances (EPS), which are a major component of biofilms, are composed of several natural polymers with negatively charged functional groups like hydroxyl (OH^-) and sulfate (SO_4^{2-}), which could enhance the electrostatic attraction of lead species toward the pipe surface, but detachment of biofilm might result in an overall lower lead surface loading onto the biofilm-laden plastic pipes (Ahamed et al., 2020).

3.4. Effect of water pH on lead release from new water pipes under flow conditions

This study investigated the effect of water pH on lead release from new PEX-A and HDPE pipes that had previously accumulated lead. The lead release experiments were conducted at pH 7.8, 6.0, and 5.0 under flow conditions. At pH 7.8, there was no detectable lead release from either of the new plastic pipes, indicating that there was an insignificant physical detachment of the lead species into the flowing water. However, when the new PEX-A pipes were exposed to a clean synthetic tap at a lower pH of 6.0, only 1% (5.9 μg) and 0.5% (3.5 μg) of the initially accumulated lead were released to the contact water after 5 min and 2 h of exposure, respectively. For new HDPE pipes, this condition resulted in only releasing 0.5% of the initially accumulated lead after 5 min exposure to the clean synthetic tap water. The released lead species were found to be reaccumulated onto the plastic pipes, and no more lead release was detected by longer exposure of the plastic pipes to clean synthetic tap water at pH 6.0. With a further reduction of water pH to 5.0, the plastic pipes exhibited an increased lead release that continued to increase over the 5 d release period (Fig. 3). The percentage of lead mass released by PEX-A pipes increased from 1% to 4.5% by increased exposure duration from 5 min to 5 d, at pH 5.0.

By increasing the release period from 5 min to 2 h, the lead release from new HDPE pipes was significantly reduced (p-value<0.05) from 2.7% to 0.8%. This indicates that exposing lead-accumulated HDPE pipes to lead-free synthetic tap water under flow conditions led to a rapid release of loosely adhered lead species to the HDPE surface. However, over time, some of these released lead species were redeposited onto the pipes. At pH 5.0, 99.7% of lead exists as dissolved species $[Pb^{+2}]$, while this percentage decreased to 83.1% at pH 6.0. At the lower pH values, rather than the dissolution of lead species, H^+ ions competed with dissolved lead species to occupy available surface sites on the plastic pipes. Since H^+ (H_3O^+) ions are smaller [ionic radius of 0.3 Å] than lead species [ionic radius of 1.20 Å] (Andersson et al., 2008), they can replace the lead species that have been accumulated on the available surface sites (Andersson et al., 2008; Makris et al., 2014; Kurajica et al., 2023).

We used copper pipes as our controls and compared our metal release result at pH = 5.0 to them. The copper pipes released a significantly (p-value <0.05) lower percentage of lead (6.5%) after 5 d of the release experiment compared to the PEX-A and HDPE pipes, which released 25.5% and 18.5% of initially accumulated lead, respectively. However, the percentage of Pb mass released from copper pipes was not statistically different from PEX-A and HDPE pipes after 5 min, 2 h, and 6 h of release periods. These results were in agreement with the findings reported by Al-Malack et al. (2001) (Al-Malack, 2001) that showed increased lead migration from polyvinyl chloride (PVC) pipes into the water by pH reduction from 9.0 to 5.0 to the water. The USEPA Secondary Maximum Contaminant Level (SMCL) for pH is 6.5–8.5 for tap water. However, under real building plumbing conditions, the pH reduction of tap water may occur due to nitrification (Zhang et al., 2008a). Zhang et al. (2007) reported a pH reduction of up to 0.75 units in a PVC premise plumbing due to the nitrification process (Zhang et al., 2008b). The extent of pH reduction may vary based on the extent of nitrification and the initial alkalinity of tap water (Zhang et al., 2009).

3.5. Lead release from new and biofilm-laden water pipes under stagnant conditions

A significantly greater level of lead was released from biofilm-laden PEX-A pipes compared to new PEX-A pipes (p-value<0.05), as shown in Fig. 4. During the 5 d release experiment, the mass of lead released from new PEX-A pipes increased from 0.02% (0.1 μg) after 5 min to 0.40% (2.2 μg) (p-value<0.05). The maximum release (3.3 μg, 0.5%) occurred after 12 h of exposure to stagnant, clean synthetic water. In contrast, the mass of lead released from the biofilm-laden PEX-A pipes increased from 1.7% (9.8 μg) after 5 min to 4% (21.3 μg) after 5 d (p-value<0.05), which was 10 times greater than that released from the new PEX-A pipes (p-value<0.05). Additionally, the instantaneous Pb release (5 min) from new PEX-A pipes under stagnant conditions was very small (0.1 μg) compared to the biofilm-laden PEX-A pipes (10 μg) (p-value<0.05). Similar to PEX-A pipes, the biofilm-laden HDPE pipes released a significantly greater level of lead (p-value<0.05) compared to new pipes (Fig. 4). The lead release from the new HDPE pipes increased from 0.2 μg ($<0.05\%$) after 5 min to 3.8 μg (0.7%) after 5 d exposure to clean synthetic tap water at pH 5.0 (p-value<0.05). However, the presence of biofilm on the new HDPE pipes increased the lead release from 5.7 μg (1%) after 5 min to 31.0 μg (5%) after a 5 d release period (p-value<0.05). The instantaneous percentage of lead release (5 min) from biofilm-laden HDPE pipes was found to be 1%. However, it was negligible for new HDPE pipes. The lead release from new copper pipes

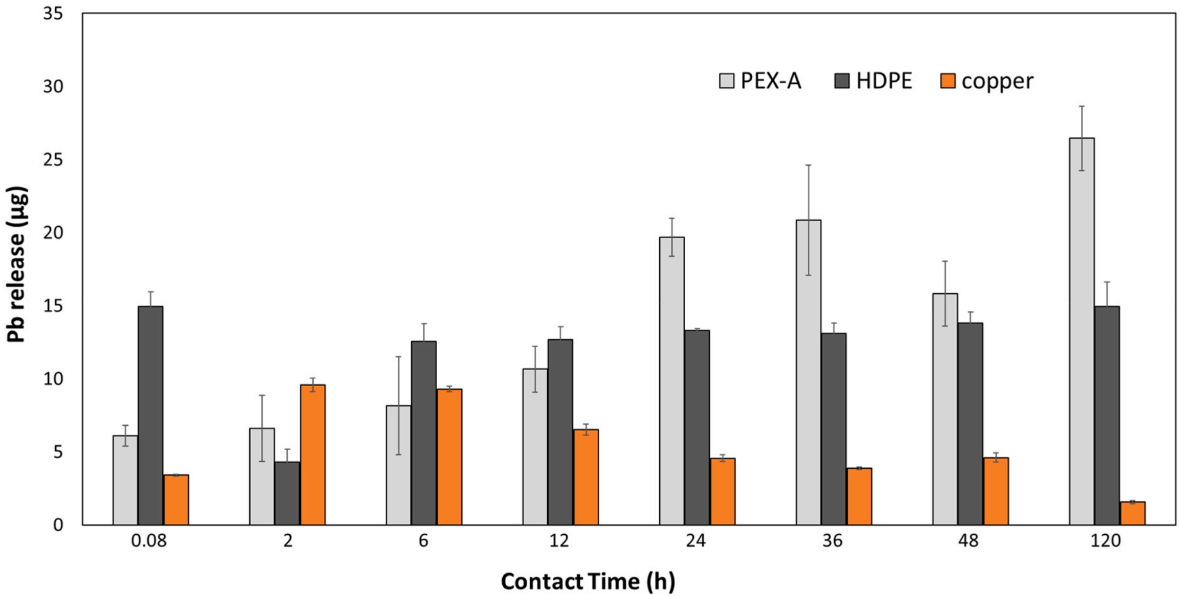


Fig. 3. The mass of Pb released from 1 ft of new PEX-A, HDPE, and copper pipes under flow condition at pH = 5.0. Error bars represent standard deviation (n = 3).

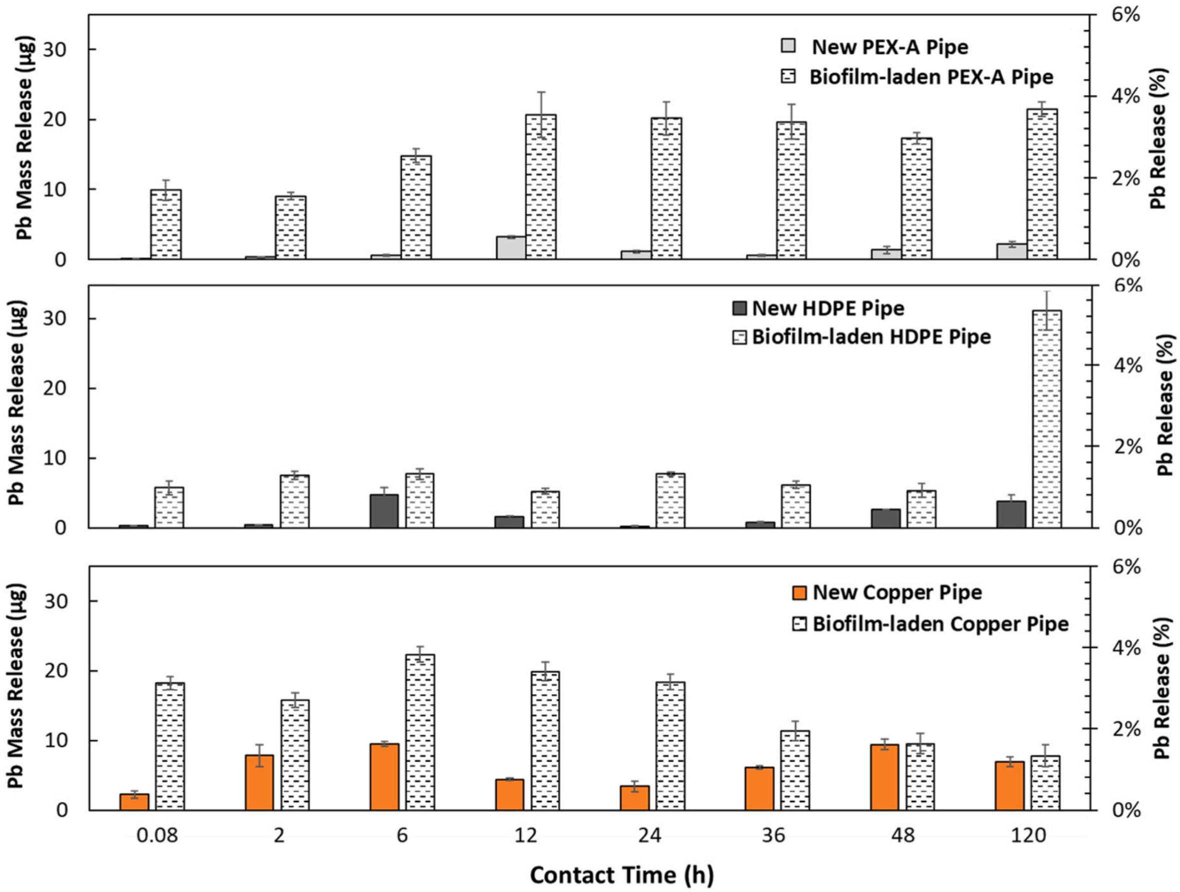


Fig. 4. The mass of Pb released from 1 ft of new PEX-A, HDPE, and copper pipes under stagnant conditions at pH = 5.0. Error bars represent standard deviation (n = 3).

increased from 2.2 µg (0.40%) after 5 min to 7 µg (1.2%) after 5 d of exposure to clean synthetic water (p-value<0.05). However, the biofilm-laden copper pipe released 18.2 µg (3.2%) of its accumulated Pb after 5 min, which was 8-fold higher than the mass of lead released from a new copper pipe (2.2 µg) (p-value<0.05). Under stagnant conditions, after 120 h of lead release period, the lead release from three different types of new pipes was significantly different (p-value<0.05) and followed the order of copper > HDPE > PEX-A. At a similar time period, lead release from biofilm-laden pipes was also significantly different (p-value<0.05) and followed the order of HDPE > PEX-A > copper.

By comparing the results obtained after a 5 d exposure period under stagnant conditions, we found that the greatest percentage of lead released from biofilm-laden HDPE pipes (5.3%), followed by biofilm-laden PEX-A (3.7%) and biofilm-laden copper pipes (1.4%). The accessibility of adsorbed lead species to the contact water and the strength of metal-surface site attachment could have influenced the extent of lead release (Salehi et al., 2021). At pH 5.0, that release experiments were conducted, the concentration of H^+ present in the water was 631 orders of magnitude greater than in water with pH 7.8, where the metal accumulation experiments were conducted. Moreover, considering the Pb speciation results, the Pb species [e.g., $Pb_3(PO_4)_2$] precipitated onto the pipes during the accumulation experiments. However, exposure to water at pH = 5.0 could have resulted in their dissolution. The water pH could also affect bacteria-surface interactions and, in turn, their attachment to pipe inner walls. At neutral pH, most of the biofilm-forming bacteria have a net negative surface charge due to the presence of anionic groups (e.g., carboxyl and phosphate) on cell surfaces. Thus, electrostatic repulsion could take place upon their interaction with negatively charged pipe surfaces (Maity et al., 2021). Our recent research showed that the zeta potential of new and biofilm-laden PEX-A pipes varies from 24 mV to −62 mV and from −71 mV to −111 mV, respectively, when pH increases from 5 to 7.8 (Haddiuzaman et al., 2023). The zeta potential of new and biofilm-laden HDPE pipes varies from −52 mV to −110 mV and from −78 mV to −118 mV, respectively, when pH increases from 5 to 7.8 (Hadiuzzaman, 2023). The schematic illustrating the lead accumulation and release process for plastic pipes is shown in Fig. 5. The pH reduction can also contribute to the biofilm detachment by altering the chemical properties of the biofilm matrix (Hadiuzzaman, 2023). When the pH decreases, the chemical environment becomes more acidic. This change in pH can disrupt the electrostatic interactions within the biofilm matrix. Increased concentration of H^+ ions at lower pHs causes more protonation of the surface groups, reduces the negative surface charge, promotes the surface hydrophobicity, and thus impacts the binding strength of bacterial colonies to the substrate (Maity et al., 2021). Biofilm maturation could contribute to the Pb release from pipe surfaces by limiting the oxygen transfer in thick biofilms. As biofilms mature, they become more complex and thicker, with greater stratification of microbial communities and extracellular polymeric substance (EPS) layers. As a result, facultative aerobic bacteria become anaerobic and convert organic substrates into volatile fatty acids and insoluble gases, both of which may serve to weaken the biofilm structure (Krasowska and Sigler, 2014). This complexity can create microenvironments within the biofilm that are less accessible to nutrients and oxygen, leading to the formation of anaerobic zones. The anaerobic conditions can result in the production of organic acids and other metabolites that can damage the biofilm matrix and promote detachment (van Loosdrecht et al., 1989).

3.6. Lead release from new and biofilm-laden water pipes under flow conditions

There was a sudden decrease in the mass of Pb released from new PEX-A pipes under flow conditions after 48 h (Fig. 6). Therefore, biofilm-laden PEX-A pipes released a lower mass of Pb (4.5 μg) compared to new PEX-A pipes after 5 d of the release experiment (p-value$<$0.05). The lead release by new HDPE pipes was almost constant during the 5 d exposure period except for the 2 h time interval. Similarly, biofilm-laden HDPE pipes released 6.6 μg Pb after 5 min, which reduced to 3.8 μg after 5 d (p-value$>$0.05). No significant difference was found in the mass of lead released from new (1.6 μg) and biofilm-laden copper pipes (2.0 μg) after 5 min exposure to clean synthetic water (p-value$>$0.05). The lead mass released from biofilm-laden copper pipes was 2.0 μg (0.35%) after 5 min and increased to 9.6 μg (4%) after 2 h exposure to clean synthetic tap water (p-value$<$0.05). After 6 h, there was a descending trend in lead mass released from biofilm-laden pipes under flow conditions. At pH 5.0, the presence of biofilm resulted in a significant increase in the release of lead from plastic and copper pipes under flow water conditions compared to the stagnant water condition. This may be due to a greater concentration gradient created by flowing water. Under flow conditions, after 120 h of the lead release period, the lead release from three different types of new pipes were significantly different (p-value$<$0.05) and followed the order of copper > HDPE > PEX-A, which was similar to the order identified under stagnant conditions. At a similar time period, lead release from biofilm-laden pipes was also significantly different (p-value$<$0.05) and followed the order of HDPE > PEX-A and copper.

3.7. Limitations and implications

In this study, the biofilms were developed onto the pipes only for

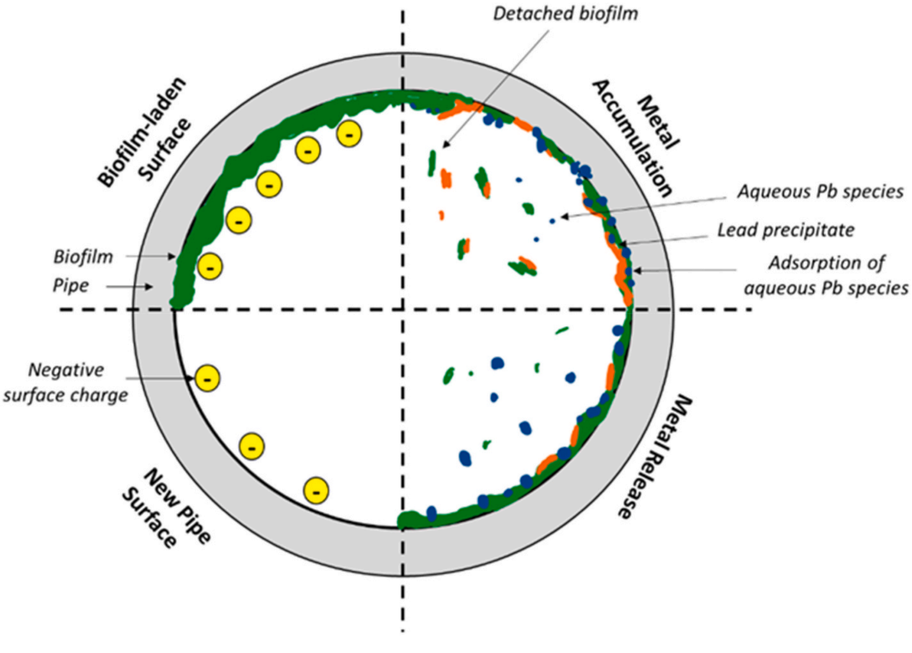


Fig. 5. The schematic illustrating the mechanisms of lead accumulation onto and release from new and PEX-A pipe.

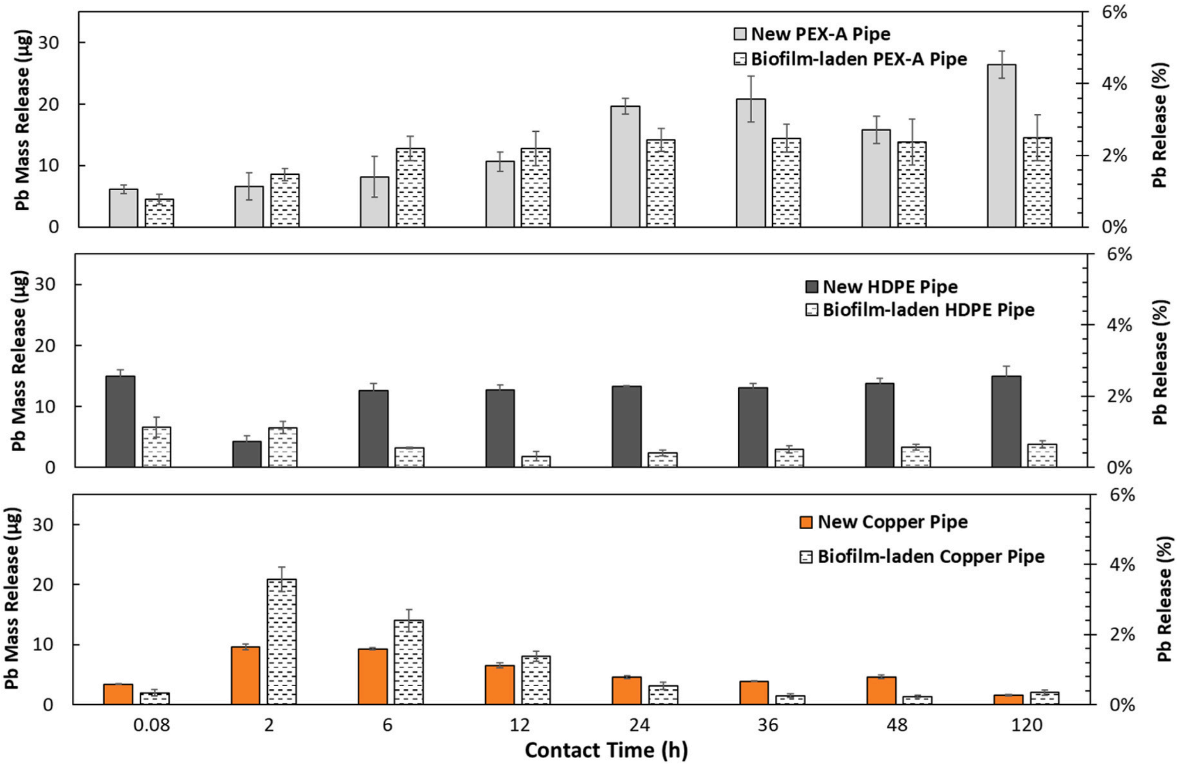


Fig. 6. The mass and percentage of Pb released from 1 ft of a) PEX-A, b) HDPE, and c) copper pipes into 1 L of synthetic tap water at pH 5.0 under flow conditions. Error bars represent standard deviation (n = 3).

three months. However, in household plumbing conditions, biofilms can accumulate for several years. Consequently, the thickness, microstructure, biological composition, and adhesion of these biofilms to pipe surfaces may differ from those observed in our short-term study. These differences could potentially affect both metal accumulation and release in plumbing systems. Therefore, future research is needed to examine the metal release behavior of potable water pipe sections collected from the field. Furthermore, this study did not consider the impact of disinfectant residuals on lead release behavior; oxidation of lead species by disinfectant residuals could influence their dissolution in water. Hence, further investigation is necessary to unravel the critical role disinfectant residuals might play over long-term release periods. Synthetic tap water was used to conduct the metal exposure experiments in this study; however, it may not reflect the wide range of water chemistry characteristics of finished ground or surface waters. Moreover, in this study the uniformity of the biofilm formation on pipe surfaces was not investigated. However, it should be noted that biofilms are not homogeneous but rather have dynamic 'lifecycles' that include planktonic, cluster colonies, detachment, and reattachment, where these EPS amalgams detach and reattached to the surface of the pipe (or any surface) in a stochastic manner. So, there may not be a uniform growth of biofilms on the surface of the pipes, but each independent replicate pipe should have very similar growth patterns that facilitate comparisons.

Despite the limitations, the knowledge developed in this study provides a fundamental understanding of heavy metals transport within building potable water plumbing materials. Among various heavy metals that may be released into drinking water, lead exposure via drinking water remains a public health concern. It was reported by the Natural Resources Defense Council (NRDC) that between 2018 and 2020, 3 million people were served by 372 drinking water systems that had over 530 health-based violations for lead (Fedinick, 2023). Despite the absence of lead in plastic pipes themselves, lead can enter the plastic potable water plumbing systems through multiple ways, such as lead-based solder, lead service lines, or lead-containing brass fittings. As a result, when water flows through these sources, it can remove lead species from lead-containing plumbing components, which may accumulate on the inner surfaces of plastic pipes. Fluctuation in water chemistry [e.g., pH] and exposure to elevated water flow rates could release the accumulated lead back into the water. The variation of lead release from the plastic surface by water chemistry conditions highlights the role of water sources, treatment practices, and water quality fluctuations in lead mobility within building plumbing systems. This finding, along with previous literature indicating a greater biofilm detachment from plastic plumbing than copper pipes, raises concerns about the potential risk of plastic pipes to public safety compared to copper pipes. This research revealed the significant role of water chemistry fluctuations, such as pH reduction, in increasing lead release from plastic pipes by forming lead precipitates and inhibiting biofilm development on the pipe surface. Therefore, water utilities using different sources of water should develop strategies to avoid significant water pH fluctuations within their network to limit the risk of metal release into tap water.

To mitigate lead exposure in tap water, it's important to decontaminate the potable water plumbing systems after short- or long-term exposures to lead in tap water. This study advances the development of decontamination strategies to remove heavy metals that have accumulated over the years or under specific circumstances such as lead service line replacement. The decontamination practice can be conducted at the individual building that has an issue of lead contamination of its plastic potable water plumbing. The results indicated that reducing the water pH can cause loosely attached lead species to be released from the pipe surfaces into the water. However, in water stagnant conditions, lead release from biofilm-laden plastic pipes was greater than from new plastic pipes, which showed very low lead release under these conditions. On the contrary, when there was water flow, lead release from new plastic pipes was slightly greater than from biofilm-laden water pipes. Therefore, when using acidic water for metal decontamination of the plumbing system, the condition of the plumbing system should be taken into account. It should be noted that, regardless of stagnant or flow conditions, only a small percentage of accumulated lead species, mainly the loosely attached ones, were released into the water. This suggests that typical fluctuations in tap water chemistry might not result in a significant release of accumulated lead to the tap water. However, in the future, the use of chemical decontamination agents should be further investigated for more effective decontamination of metal-contaminated plastic piping materials.

4. Conclusion

Despite the emergence of plastic potable water pipes as a solution to issues associated with metallic pipe corrosion, there is still a need for more information on the heavy metals transport within these pipes. Therefore, this study was conducted to examine the impact of biofilm presence and water pH on the lead release from lead-accumulated PEX-A and HDPE pipes in comparison to lead-accumulated copper pipes under water flow and stagnation conditions. The results showed an order of PEX-A > HDPE > copper pipes for the magnitude of biofilm biomass accumulation on pipes after three months of conveying the municipal tap water. Both new and biofilm-laden PEX-A pipes accumulated greater levels of lead than HDPE and copper counterpart pipes. The reduction of water pH from 7.8 to 5.0 enhanced the lead release from all pipes, in which greater lead was released from new PEX-A pipes compared to the new HPDE and copper pipes. The comparison of lead release from lead-accumulated plastic water pipes showed a greater lead release from biofilm-laden water pipes compared to new plastic water pipes in stagnant conditions. The knowledge developed in this study could be further utilized to inform future studies on the development of decontamination practices for metal-contaminated plastic plumbing materials.

Credit author statement

Shima Ghoochani: Methodology, investigation, writing, visualization; Nahreen Mirza: Investigation; Md Hadiuzzaman: Investigation; Shawn P Brown: Methodology, Writing, and Editing; Maryam Salehi: Conceptualization, Methodology, Writing and Editing, Demonstration, Supervising.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2023.122520.

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